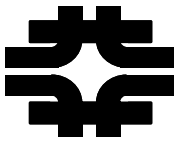

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RP. NOTE 141

Calculation of the Induced Radioactivity Levels and Hydrogen Gas Evolution in the NuMI RAW Systems

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(June 2003)

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Introduction

The NuMI[1] beam line starts at the extraction from the Main Injector, followed by a Carrier pipe, baffle and target system, two focusing horns, Decay Pipe, and the Hadron Absorber. This note describes the calculation of the induced activity and hydrogen gas in the Radioactive Water (RAW) Systems.

The interaction between the ionizing radiation and water leads to the formation of short-lived radical species (OH, H and electrons mainly) and stable molecular species (O_2, H_2O_2 and H_2). While most of the ions recombine to form water, hydrogen gas and other species are also produced. The amount of hydrogen gas produced is calculated by a simple scaling argument to assess the level of safety precautions required.

Methodology

1. Induced Activity Calculations

The buildup of specific activity, a_i, of a particular radionuclide, i, as a function of irradiation time, t_{irrad} (sec), in a mixture of chemical elements is calculated using,

$$a_i(t_{irrad}) = \sum_j n_j \sigma_{ji} \phi \left[1 - \exp(-\lambda_i t_{irrad})\right] \qquad (\text{Bq cm}^{-3}). \qquad (1)$$

Since this note deals with the activation of pure water, only the production of different isotopes from oxygen is considered (j=*1)*. Production of ^{3}H from natural hydrogen is a two-step neutron capture process with a much smaller cross section, which will be ignored here. In Eq. 1, it is assumed that the water is exposed to a uniform flux density of particles of a particular type or energy, ϕ ($\text{cm}^{-2}\text{s}^{-1}$), which is constant over time. Also, n_j is the number density (cm^{-3}) of oxygen atoms and σ_{ji} is the cross section (cm^2) for producing radionuclide i from the interactions of particles of a particular type or energy with oxygen. λ_i is the decay constant (s^{-1}, inverse mean life) of radionuclide i. After the irradiation has ceased, if the radioactivity is allowed to decay for a time period t_{decay}, the specific activity present will be a function of the decay time, t_{decay}, as follows:

$$a_i(t_{decay}) = a_i(t_{irrad}) \exp(-\lambda_i t_{decay}) \qquad (\text{Bq cm}^{-3}). \qquad (2)$$

The values of σ_{ji}, were obtained from the standard reference by Barbier[2] except those related to the production of ^{3}H, which was taken from the results of Konobeyev and Korovin[3]. From oxygen, only five radionuclides having lifetimes of significance are produced. Table 1 gives the cross sections used in the calculations. As Table 1 shows, the most significant radioisotopes, from a contaminant aspect are ^{3}H, and to a lesser

extent ^{7}Be, because of their rather long half lives. However, from a dose rate from a RAW system point of view, tritium dose not contribute, because of its very low energy decay beta particle. Only 10% of ^{7}Be decays are via gamma emissions, while ^{11}C, ^{13}N and ^{15}O are positron emitters producing 0.511MeV gamma rays. Therefore, allowing cooling time before getting near a RAW system is a very effective way of reducing the dose rates.

Table 1. Radionuclides with half-life > 2 minute that can be produced in cooling water

Radionuclides	**Decay Constants, λ_i (s^{-1})**	**Production Cross-Section from oxygen (mb)**	**Half life**
$^{3}H^{a}$	1.78×10^{-9}	25	12.33 years
^{7}Be	1.51×10^{-7}	8	53.29 days
^{11}C	5.69×10^{-4}	10	20.385 minutes
^{13}N	1.16×10^{-3}	5	9.965 minutes
^{15}O	5.67×10^{-3}	35	122.24 seconds

The fluxes of the particles for different regions are obtained from various MARS[4] simulations of the experimet[5]. All calculations are done for a beam proton intensity or 4E20 per year.

The Fermilab Radiological Control Manual (FRCM) suggests keeping the tritium concentrations in the cooling system below 0.67 microCuries/cc. However, some of the NuMI RAW systems may exceed this limit due to high particle fluxes or it may not be feasible to replace the cooling water frequent enough to keep the concentrations below the FRCM value. For these cases, multi-tier containment methods can be used, with the proper operational precautions during the handling of the RAW systems. When possible the high tritium concentration water is disposed of as solidified low-level radioactive waste.

2. Radiolysis of water

Calculations are based on the G-number, which is the number of atoms/ion produced of a given kind, per 100 eV of ionizing energy. The exact value depends on several factors, and radio-chemists prefer to simulate (i.e. with Monte Carlo) the exact setup. Ranges are from 0.1 to 5. Based on the ionization energy of water molecule and the extant information on the operation of the anti-proton source and measurements at SLAC[6], a value of 0.2 was used for these calculations. A 4% hydrogen gas mixed in air is considered to be explosive. If well mixed with the air, the hydrogen concentrations will not get to 1%. However, most of the hydrogen is in the closed loop RAW system. If released, because of its tendency to accumulate at the high places near the ceiling doom, or gaps and depression, it is possible to form explosive mixtures. A mitigating method will be designed into the system wherever the calculation predicts significant hydrogen gas production

Results and Discussions

1- Target

The NuMI target is composed of forty-seven 20 mm long graphite segments, separated by 0.3 mm. Each segment is 6.4 mm wide and 18 mm high. Target is cooled by water circulating through two tubes, running along the length of the target and back. The tubes are 1.07 cm from the center of the target. The inner diameter of a tube is 5.4 mm. The amount of water present in the tubes is very small. The flux of the particles in the cooling water is assumed to be ten times that in the area between the conductors of the first horn[6]. One-year operation will build to about 3.5 Curies of activity, out of which only 0.8 Curies is the long-lived isotope tritium.

The dose rate at one foot from two cooling lines was calculated by modeling the system as a shielded line source, where the thickness of the shield is the thickness of the tubes (1.6 mm or 1/16 in.).

$$\Phi(rad/hr) = \frac{S_L k}{2\pi d} F(\theta, b_1) \qquad (3)$$

Where S_L is activity per unit length, d is the distance from the line source, k is a factor that converts activity to dose rate. k is a function of photon energy and the gamma ray absorption mass coefficient. Angle θ is the half angle subtended by the line source and b_1 is the shield thickness expressed in mean free path lengths. F is called the secant integral function or the Sievert integral, which is computed numerically.

The absorbed dose rate at one foot from these lines, without any cooling time allowed, is 14.3 rads/hr. Only 0.2 rad/hr is from ^{7}Be, the rest is due to the short-lived isotopes. Since, the target itself is expected to produce a 6000 rad/hr radiation field, any shielding used for the target will shield the cooling lines as well. The level of the activity of the raw system will require some containment measures, to protect the personnel and possibility of leakage into the ground.

For the radiolysis calculations, the energy deposition in the graphite target was scaled to the density of water. The results show that about 1 cm^3 of hydrogen gas is produced per day, which is insignificant in the large volume of air (about 5E7 cm^3 around the baffle target and horn 1) present.

2- The Focusing Horns

The focusing horns are made of double parabolic, nickel plated aluminum inner conductor and an anodized aluminum outer conductor. Water spray on the conductors will remove the generated heat. It is assumed that there is about 2 liters of water between the two conductors at any time[6]. This water collects in the retention tanks under the horn and then returns to the surge/accumulation tank. It was assumed that there is no more than 20 gallons of water in the retention tank and 100 gallons in the accumulation tank[6].

Activation of the Cooling Water

The average of the MARS predictions of the particle fluxes in the inner and the outer conductors was used for the calculation. As tables 2 and 3 show, most of the activity is due to the short-lived isotopes. However, for the disposal of this RAW system, tritium is the important isotope. After one year of operation, there are about 6.8 Curies of tritium in Horn1 and 2.4 Curies in Horn2.

Table 2. Induced activities in Horn1 RAW system.

After 1 year operation	Between Horn1 Conductors	Horn1 Retention Tank	**Total Activity**	**Specific Activity**		**Total Activity**	**%Activity**
	Bq	**Bq**	**Bq**	**Bq/cm3**	**(Ci/cc)**	**(Ci/Year)**	
H3	2.14E+10	4.60E+10	6.75E+10	1.78E+05	4.82E-06	1.82E+00	2.3
Be7	1.25E+11	2.67E+11	3.92E+11	1.04E+06	2.80E-05	1.06E+01	13.4
C11	1.57E+11	3.37E+11	4.94E+11	1.30E+06	3.53E-05	1.34E+01	16.9
N13	7.85E+10	1.68E+11	2.47E+11	6.52E+05	1.76E-05	6.68E+00	8.4
O15	5.50E+11	1.18E+12	1.73E+12	4.57E+06	1.23E-04	4.67E+01	59.0
				Total=	2.09E-04	7.92E+01	
		Total activity after 1 hr cooling=			3.76E-05	1.42E+01	

Table 3. Induced activities in the Horn2 RAW system

After 1 year operation	Between Horn2 Conductors	Horn2 Retention Tank	**Total Activity**	**Specific Activity**		**Total Activity**	**%Activity**
	Bq	**Bq**	**Bq**	**Bq/cm3**	**(Ci/cc)**	**(Ci/Year)**	
H3	2.93E+09	1.70E+10	1.99E+10	5.60E+04	1.51E-06	5.38E-01	2.3
Be7	1.70E+10	9.87E+10	1.16E+11	3.25E+05	8.79E-06	3.13E+00	13.4
C11	2.14E+10	1.24E+11	1.46E+11	4.10E+05	1.11E-05	3.94E+00	16.9
N13	1.07E+10	6.22E+10	7.30E+10	2.05E+05	5.54E-06	1.97E+00	8.4
O15	7.50E+10	4.36E+11	5.11E+11	1.44E+06	3.88E-05	1.38E+01	59.0
				Total=	6.57E-05	2.34E+01	
		Total activity after 1 hr cooling=			1.18E-05	4.20E+00	

Shielding of the Horns Supply/Surge tank and the Filters/DI bottles

As a simplifying assumption for this calculation it was assumed that all of the activity end up in a 100-gallon tank of 1 ft (30 cm) radius. It was further assumed that the tank is made of 1/8 in. (3.2 mm) steel. For the shielding calculations, the geometry was assumed to be that of an absorbing cylindrical volume-source with shield (tank steel wall). Then the dose rate from such a source is given by

$$\Phi(rad/hr) = \frac{BRkS_v}{\pi} G(k', p, \mu_s R, b_1) . \qquad (4)$$

Where b_1 and k are similar to the ones used for target cooling calculations. R is the radius of the source, S_v is activity per unit volume, k' is the ratio of the height of the cylinder to its radius, *p* is the ratio of the distance from the center of the cylinder to its radius, and μ_s is the mass absorption coefficient of water for photons. The non-analytical attenuation function G is again evaluated numerically. B is the point source buildup factor of gamma rays in the iron shield. It is calculated using

$$B = A_1 e^{-\alpha_1 b_1} + (1 - A_1) e^{-\alpha_2 b_1} . \qquad (5)$$

Where b_1 is the mean free path for gamma rays in iron, and
A_1= 31.379, α_1=-0.0684 α_2=-0.0374.

Table 4. Dose rates from the Horn1 and Horn2 RAW surge/accumulation tanks.

Dose rate at 1ft from the tank	Dose rate with no shielding	Dose rate after 1"Pb	Dose rate after 2"Pb	Dose rate after 3"Pb	Dose rate after 4"Pb	Fraction of Dose due to Be-7
	(Rad/hr)	(Rad/hr)	(Rad/hr)	(Rad/hr)	(Rad/hr)	
Horn1 (no cooling)	1.19E+01	2.50E-01	4.11E-03	6.22E-05	8.88E-07	0.20%
Horn1 (1hr cooling)	5.01E-01	1.05E-02	1.73E-04	2.62E-06	3.73E-08	5.50%
Horn1 (2hr cooling)	2.20E-01	4.61E-03	7.58E-05	1.15E-06	1.64E-08	32.10%
Horn1 (1 day cooling)	1.78E-01	3.74E-03	6.15E-05	9.31E-07	1.33E-08	100%
Horn2 (no cooling)	3.75E+00	7.87E-02	1.29E-03	1.96E-05	2.79E-07	0.20%
Horn2 (1hr cooling)	1.58E-01	3.31E-03	5.43E-05	8.22E-07	1.17E-08	5.50%
Horn2 (2hr cooling)	6.90E-02	1.45E-03	2.38E-05	3.60E-07	5.14E-09	32.10%
Horn2 (1 day cooling)	5.60E-02	1.18E-03	1.93E-05	2.92E-07	4.17E-09	100%

The results of calculations are given in Table 4. for different cooling times. As the last column shows, after a few hours of cooling most of the dose from the RAW system is due to ^{7}Be. Most of the ^{7}Be will be trapped in the filters, DI bottles and plate out in the pipes. The suggested lead shielding (or its equivalent) can be used for local shielding of DI bottles and filters. Other measures can be used to protect the personnel from the RAW system radiation.

Air Concentrations of the Activated Water from a Defunct Horn1 Tank

It is useful to assess the effects of not draining all of the water out of a horn that will be stored in the target hall morgue. Since Horn1 cooling water receives more radiation than Horn2, it will be used for this assessment. The tritium concentration in the Horn1 RAW system, after one year of operation is 1.8e-5 Ci/ml. Assume that 5 gallons RAW was left in the horn, which is stored in the morgue. Allowing this water to evaporate, in one week, and leak out of the morgue and using the air flow rates out of the two decay tunnel stacks of 3300 cfm (from August 2001 FESS drawing for Project 6-7-9), right on top of the stack the concentration of the tritium will be about 0.39 pCi/ml. This will further dilute through mixing and propagation in the atmosphere before it gets to the site boundary. Annual average off site will be insignificant. A radiation worker, working in the area this whole week, will receive about 7.5 mrem by breathing the entire tritium vapor in the air.

Radiolysis of the Horns Cooling Water

For the calculation of the hydrogen gas production in the volume between the conductors the MARS energy deposition calculations in the aluminum conductors was averaged and scaled down to the density of water. Energy deposition prediction in the retention tank area was not available. The energy deposition in the water between the two conductors was scaled down according to the hadron flux reduction, going from between the conductors to the retention tank area. The results of the calculations are shown in table 5.

Table 5. Hydrogen gas production in the horns based on energy deposition calculation using MARS.

Average E-deposition (GeV/g/pp)	Average E-deposition (GeV/g/pp) for water	GeV	H2 molecules	Gallons of H2/year	Gallons of H2/day	Gallons of water lost/yr	
2.76E-05	1.02E-05	8.17E+18	8.17E+24	80.2	0.2	0.03	Horn 1
7.81E-06	2.89E-06	1.75E+19	1.75E+25	172.1	0.5	0.07	Retention tank1
			TOTALS=	*252.3*	*0.7*	*0.10*	
2.37E-06	8.78E-07	7.03E+17	7.03E+23	6.9	0.0	0.00	Horn 2
1.82E-06	6.73E-07	4.08E+18	4.08E+24	40.1	0.1	0.02	Retention tank2
			TOTALS=	*47.0*	*0.1*	*0.02*	

As mentioned in the introduction the amount of hydrogen gas produced in the horns RAW systems is significant. Thus, mitigation is being designed into the RAW systems.

3- Decay Pipe

The NuMI Decay Pipe has an inner diameter of 1.98 m and is 677.1 m long. Twelve 1"-diameter copper, water cooling lines, uniformly distributed around the circumference of the pipe, along the length of the Decay Pipe. The total volume of the cooling lines is about 640 gallons and the supply tank's volume is 724.5 gallons. The MARS calculation has subdivided the Decay Pipe into 270 segments and flux for each region is given. The calculation of the induced activity was done for each segment separately and the resulting activities were summed. The results of the calculations are given in Tables 6 and 7.

Table 6. Calculated activities, with and with out cooling.

	H3	Be7	C11	N13	O15
Specific Activity (Ci/cc)	2.38E-09	1.38E-08	1.75E-08	8.73E-09	6.11E-08
Total uncooled (%)	2.30%	13.38%	16.86%	8.43%	59.02%
Total cooled for 1hr (%)	12.81%	74.37%	12.10%	0.72%	0.00%

Table 7. Calculated dose rates with and with out cooling, shielding of the supply tank, and the volume of the hydrogen produced.

Total Dose rate at the tank after 1 year of operation and No cool down=	14.82	(mrem/hr)
Dose after 1" of Lead =	0.31	(mrem/hr)
Total Activity in the water after 1 year of operation and No cool down=	283.90	(mCi/year)
Total Dose rate at the tank after 1 year of operation and 1hr of cool down=	0.62	(mrem/hr)
Dose after 1" of Lead =	0.01	(mrem/hr)
Total Activity in the water after 1 year of operation and 1hr of cool down=	51.03	(mCi/year)
Hydrogen Gas Production =	6.1	(gallons/year)

As Table 6 shows, most of the un-cooled activity is due to the ^{15}O, which has a two-minute half-life. One hour of decay time will reduce the total activity of the RAW significantly. After one year of operation, only 6.5mCi of ^{3}H and 38mCi of ^{7}Be is produced, the rest are short-lived isotopes. Since the tritium in the tank does not produce any dose rates outside the tank, and most of the ^{7}Be is trapped in the filters and the DI bottle, the amount of shielding shown in table 7 is an upper limit, and may apply only to the filters and the DI bottle. No significant shielding is required for the Decay Pipe RAW system. The amount of the hydrogen produced in the RAW system is also relatively small, and daily purging and venting with small amounts of helium gas should be sufficient.

4- Hadron Absorber

The Hadron Absorber is placed at the end of the Decay Pipe to absorb all particles except the neutrinos and a fraction of the muons. The core of the Hadron Absorber consists of 8 aluminum modules followed by 10 layers of Continuous Cast Stainless Steel (CCSS). Each module has, for redundancy, two cooling loops. The length of the aluminum passages in the core is 17.3 feet and the length of the lines behind the core is 17 feet. The combined volume is 25 gallons, with 12.6 gallons in the core and 12.4 gallons in the lines behind the core. The lines behind the core are the lines that run past the CCSS. The total volume of the tank is 135.3 gallons[8]. Star densities used for the calculation of the induced activities in the aluminum and CCSS loops were obtained from NuMI note 779[9]. All previous calculations of the Hadron Absorber used the star densities from the 1999 NuMI note 727[10]. The results of the calculations for one year of operation are shown in Tables 8,9 and 10.

Table 8. Induced activities in the Hadron Absorber RAW system.

After one year operation	Total Activity	Specific Activity		Total	% Activity
	Bq	Bq/cm3	(Ci/cc)	(Ci/Year)	
H3	3.56E+08	6.95E+02	1.88E-08	9.62E-03	2.3%
Be7	2.07E+09	4.04E+03	1.09E-07	5.59E-02	13.4%
C11	2.61E+09	5.09E+03	1.38E-07	7.05E-02	16.9%
N13	1.30E+09	2.54E+03	6.88E-08	3.52E-02	8.4%
O15	9.12E+09	1.78E+04	4.81E-07	2.47E-01	59.0%
		Total=	8.16E-07	4.18E-01	
	After 1 hr cooling Total=		1.47E-07	7.51E-02	

The important activities are due to tritium and ^{7}Be. The built up concentration of tritium after one year is below the FRCM recommended limit. With proper precautions, there is no need to replace this water with fresh water during the life of the experiment. The dose rate from the tank with out a cooling period is mostly due to the short-lived ^{15}O, as shown in the last column of Table 8.

Table 9. Shielding of the Hadron Absorber RAW System

After one year operation	Dose at 1ft from tank	Dose after 1"Pb	Dose after 2"Pb	Dose after 3"Pb	Dose after 4"Pb	DOSE (1hr cool.)
	(R/hr)	(R/hr)	(R/hr)	(R/hr)	(R/hr)	%
H3	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0
Be7	2.26E-03	4.74E-05	7.79E-07	1.18E-08	1.68E-10	36
C11	2.94E-02	6.17E-04	1.01E-05	1.53E-07	2.19E-09	60
N13	1.47E-02	3.08E-04	5.07E-06	7.66E-08	1.09E-09	4
O15	1.03E-01	2.16E-03	3.55E-05	5.37E-07	7.66E-09	0
Total=	1.49E-01	3.13E-03	5.14E-05	7.78E-07	1.11E-08	
Total (1hr cool.)=	6.27E-03	1.32E-04	2.16E-06	3.27E-08	4.67E-10	

The Hadron Absorber tank and controls are located in the nearby bypass tunnel. As Table 9 shows, if a 1-hour cooling period before access is not possible, some small amount of shielding may be necessary. Note that the amount of ^{7}Be produced after one year of operation is not enough to require shielding for the removed DI bottles.

Table 10. Hydrogen production in the Hadron Absorber RAW system

Cooling Loop	Gallons of H2 gas/year	Gallons of H2 gas/day	Gallons of water lost/yr
Aluminum core	2.4	6.7E-03	9.77E-04
Steel	2.4	6.6E-03	9.66E-04
TOTALS=	4.8	1.3E-02	1.94E-03

As given in Table 10, the amount of hydrogen produced due to the operation of the Hadron Absorber RAW system is very small. A simple vent to the exhaust stacks should be sufficient to purge the tank. The amount of water loss due to radiolysis is insignificant.

5- Tritium Concentration of discharge to the surface waters due to a leak in the Hadron Absorber or the Decay pip RAW System

The total volume of the aluminum core and the steel cooling water is about 25 gallons. After one whole year of operation, the concentration of tritium in the cooling water is about 18800 pCi/ml. It is assumed that all the water from the cooling lines spills, before the sensors shut the flow. Taking a tunnel inflow rate of 150 GPM. In two minutes mixing with the inflow, the concentration of the water discharging to the surface will be less than 1450 pCi/ml. This is below the surface waters discharge limit[11] of 2000 pCi/ml. Therefore; the effect of such a transient scenario will be minute.

The concentration of the tritium in the Decay Pipe RAW system is about a factor of 8 smaller than Hadron Absorber's concentration. Given the length of the decay tunnel, the mixing of the spilled water with the inflow will results in much smaller concentrations than that from hadron absorber's RAW spill.

Conclusion:

The RAW systems activity is very high during the times that the beam is on. Personnel can be protected through the use of one or combinations of; time, distance and shielding. Time could be either allowing a reasonable cooling time or/and reducing the access time. The prediction of the actual amount of ^{7}Be trapped by the filters and the DI bottle, depends on the pipe, filter and DI bottle material, the surface properties, the water metal interface, amount of suspended impurities that will be built up in the cooling water over time, the temperature of the water, etc. Due to lack of a clear operational history of the DI bottles, an operational rule of thumb is not available. However, an efficient solution would be to use the calculated lead (or equivalent) shielding for the DI bottles and filters, where a lot of ^{7}Be is expected to be trapped. The horns RAW systems, because of the high amounts of tritium buildup, should have multi-tier spill protection/containment systems, to prevent against tritium uptakes or spread of contamination. The produced hydrogen gas cannot be allowed to accumulate in the RAW system. Provisions are made to use a continuous flowing purging gas to remove and safely exhaust the hydrogen gas Up the NuMI stacks.[12] The hydrogen issue has also been discussed with Jim Priest, the Fermilab Fire safety expert. Jim Priest concurs with the hydrogen hazard assessment and its mitigation.[13]

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